PRECIPITATION SCAVENGING IN A COUPLED CHEMISTRY/CLIMATE MODEL OF SULFATE AEROSOL

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1. INTRODUCTION

Knowledge of the spatial distribution of sulfate aerosol over the globe is important because sulfate aerosols affect global climate directly by scattering solar radiation and indirectly by altering the cloud drop size distributions. The resulting negative climate forcing due to scattering by aerosols counteracts the warming associated with increased greenhouse gas concentrations; however, the pattern of forcing is quite different because the distribution of sulfate aerosol is regionally inhomogeneous (Charlson, et al., 1992; Kiehl and Briegleb, 1993). Furthermore, changes in cloud drop size distributions induced by aerosols can change the back-scattering of solar radiation by clouds and the cloud life cycle, thereby affecting global temperature and precipitation patterns.

We have previously examined the climate effects of sulfate aerosols and greenhouse gases using the Lawrence Livermore National Laboratory tropospheric chemistry model (GRANTOUR) in conjunction with CCM1 and concluded that representation of the regional distribution of atmospheric aerosols was essential for reliable prediction of climate change (Taylor and Penner, 1993). Using CCM1 meteorology, GRANTOUR has also been use to simulate the global transport and deposition of ^{222}Rn and ^{210}Pb (Dignon, et al., 1993), O_3 and OH (Atherton, 1993; Atherton et al., 1993; Penner et al., 1993a), organic nitrates (Atherton, 1989), anthropogenic aerosols (Taylor and Penner, 1993), smoke (Ghan et al., 1988), soot aerosols from biomass burning (Penner et al., 1991b), and black carbon (Penner et al., 1993b). We have also used GRANTOUR to simulate the global nitrogen budget (Atherton et al., 1991; Penner et al., 1991a) and the global sulfur cycle (Erickson et al., 1991).

Recently we have coupled GRANTOUR with the ECHAM global climate model. This provides several enhanced capabilities in the representation of aerosol interactions compared to the previous simulations with GRANTOUR coupled to CCM1. ECHAM includes a specific representation of liquid water in large-scale clouds that permits a parameterization of wet phase gas-to-particle conversion of SO_2 to sulfate. It also includes a large-scale precipitation module that we are using to improve the parameterization of scavenging by these clouds. The convective parameterization (Tiedke, 1989) includes a precipitation component that can be used to represent scavenging of aerosols by convective precipitation.

2. COUPLED MODEL

The current ECHAM/GRANTOUR model is only coupled in one direction. ECHAM is used as a meteorological driver for GRANTOUR. This has the advantage of allowing us to run ECHAM once to generate the meteorological data and then developing parameterizations for GRANTOUR with a constant set of meteorology. It has the disadvantage of not allowing feedback of the aerosols on the evolution of the weather and climate system.

We generated one year of meteorological data from ECHAM at T21 resolution. Four hour averages of the 12 variables in Table 1 were saved and constitute the meteorological data set; all are three dimensional variables defined on the ECHAM grid. GRANTOUR interpolates the variables to a constant sigma vertical coordinate that corresponds with the ECHAM variable grid for a surface pressure of 10^5Pa. Both models use the same horizontal grid. Vertical velocity is derived from U and V using the continuity equation. The parameterizations of large-scale scavenging and of convective mixing and scavenging and are actually performed on the ECHAM vertical grid with trace species mixing ratios interpolated from the GRANTOUR grid.

In the future we will provide four hour average trace species mixing ratios and/or aerosol optical properties to

ECHAM. This will permit us to evaluate some of the feedback effects of sulfate aerosols.

The aerosol precipitation scavenging parameterizations we have developed for GRANTOUR depend on the representations of the corresponding precipitation processes in ECHAM and the variables we have saved. The parameterizations are simple extrapolations of the ECHAM precipitation processes based on the assumption that sulfate aerosols are soluble, and therefore, when liquid water is present, all the sulfate mass is associated with cloud drops and precipitation.

3. LARGE-SCALE CLOUD SCAVENGING

Large-scale or stratiform precipitation in ECHAM is represented by a modified version of Sundquist's (1978) formulation (Deutsches Klimarechenzentrum, 1993). From the ECHAM simulation we save the large-scale precipitation rate, P_{LS}, and large-scale cloud fraction, F_c, and we assume all aerosol in the cloudy region is in cloud droplets. The calculation works from the top of each atmospheric column down. If P_{LS} increases downward there must have been accretion of cloud droplets and scavenging of aerosol. If we assume a cloud drop size distribution we can define an accretion (scavenging) rate as a function of P_{LS}. For simplicity we used a Marshall-Palmer size distribution,

$$n(D) = n_0 \exp^{-\Lambda D} \qquad (1)$$

where D is drop diameter, n_0 is a constant parameter, and Λ is the mean drop diameter. We specified the drop terminal velocity, V_t, with the simple expression,

$$V_t(D) = kD^{1/2}. \qquad (2)$$

Then the precipitation rate, P_{LS}, is given by

$$P_{LS} = \frac{n_0 \pi \rho_l k}{6} \frac{\Gamma(4.5)}{\Lambda^{4.5}} \qquad (3)$$

and the rate of accretion is

$$\lambda = \frac{E n_0 \pi k}{4} \frac{\Gamma(3.5)}{\Lambda^{3.5}} \qquad (4)$$

where ρ_l is air density and E is the collection efficiency. Using standard values for the constants of

$$\begin{aligned} E &= 1 \\ n_0 &= 10^7\ m^{-4} \\ k &= 130\ m^{1/2} s^{-1} \\ \rho_l &= 10^3\ kg\, m^{-3} \end{aligned} \qquad (5)$$

gives the necessary relationship between P_{LS} and λ,

$$\lambda\ [hr^{-1}] = 1.94\ P_{LS}[mm/hr]^{\frac{7}{9}}. \qquad (6)$$

Table 1. ECHAM Variables Passed to GRANTOUR

Name	Variable	Units
U	Zonal Velocity	km/hr
V	Meridional Velocity	km/hr
T	Air Temperature	K
q_v	Water Vapor Mixing Ratio	
q_l	Liquid Water Mixing Ratio	
P_{LS}	Large-Scale Precip. Rate	cm/hr
P_{CV}	Conv. Precip. Production Rate	cm/hr
M_u	Convective Mass Flux Up	kg/m^2/s
M_d	Convective Mass Flux Down	kg/m^2/s
P_h	Half-Level Pressure	Pa
F_c	Large-Scale Cloud Fraction	
K_z	Vertical Diffusion Coefficient	m^2/s

In time dt [hr], the amount of aerosol scavenged in layer j, S_{aj}, is given by

$$S_{aj} = (1 - \exp^{-dt\lambda})\ F_{cj} \chi_{aj} \frac{\Delta p_j}{g}, \qquad (7)$$

where χ_{aj} is the aerosol mixing ratio, Δp_j is the pressure thickness of layer j, and g is the acceleration of gravity.

If P_{LS} decreases downward, there must have been evaporation of rain and possible resuspension of aerosol. The fraction of rain that evaporates in layer j, f_e, is

$$f_e = \frac{P_{LS(j-1)} - P_{LS(j)}}{P_{LS(j-1)}} \qquad (8)$$

(j increases downward). Since most of the evaporated water comes from drops that only partially evaporate and do not resuspend aerosol, we assume that the fraction of rain in drops that evaporate completely, f_{ec}, is given by

$$f_{ec} = f_e^n \qquad (9)$$

where n is currently 2. Therefore, the amount of aerosol resuspended in layer j is

$$R_{aj} = f_{ec} A_{j-1} \qquad (10)$$

where A_{j-1} is the amount of aerosol in rain in layer j-1.

Working from the top down

$$A_j = A_{j-1} + S_{aj} - R_{aj} \qquad (11)$$

and

$$\chi_j^{(t+dt)} = \chi_j^{(t)} - (S_{aj} - R_{aj}) \frac{g}{\Delta p_j}. \qquad (12)$$

4. CONVECTIVE CLOUD SCAVENGING

The parameterization of scavenging by convective clouds in GRANTOUR is a part of the convective mixing algorithm just as in ECHAM. We save the rate of formation/evaporation of convective precipitation from ECHAM and have available in the parameterization the upward and downward fluxes of aerosol (all of which is assumed to be in cloud water because the convective updrafts are saturated). If a layer has an increase in convective precipitation, aerosol is assumed to be accreted at the same rate as cloud water, G_P, which is given in ECHAM by

$$\overline{\rho} G_P = K l M_u \qquad (13)$$

where ρ is air density, K is a constant, 2×10^{-3} m^{-1}, l is the cloud water mixing ratio in the convective updraft, and M_u is the updraft mass flux. Therefore the accretion rate of aerosol, G_a, is

$$\overline{\rho} G_a = K \chi_l M_u \qquad (14)$$

where χ_l is the mixing ratio of aerosol in cloud water. The upward mass flux of aerosol in cloud water is given by

$$\frac{\partial}{\partial z}(M_u \chi_l) = E_u \chi_e - D_u \chi_l - \overline{\rho} G_a \qquad (15)$$

where E_u and D_u are the entrainment and detrainment rates for the convective updraft and χ_e is the aerosol mixing ratio in the environment. Using (14) in (15) the new mixing ratio for layer j due to accretion of aerosol by rain can be approximated by

$$\chi_l^{t+dt} = \chi_l^t \frac{1}{1 + K\Delta z} \qquad (16)$$

and the convective scavenging rate, S_c, by

$$S_c = \frac{K\Delta z}{1 + K\Delta z} M_u \chi_l. \qquad (17)$$

Resuspension of aerosol in convective precipitation due to evaporation of rain in convective downdrafts is parameterized by assuming the rate of resuspension is equal to the evaporation rate. If f_{ce} is the fraction of rain evaporated, then the new mixing ratio of aerosol in the convective downdraft, χ_d, is given by

$$\chi_d^{t+dt} = (1 + f_{ce}) \chi_d^t \qquad (18)$$

and the rate of resuspension, R_c, is

$$R_c = f_{ce} \chi_d M_d. \qquad (19)$$

5. SIMULATION RESULTS

Results from the simulations will show the global distribution of sulfate scavenged by large-scale and convective clouds and the spatial distributions of suspended sulfur species. We also plan to calculate the climate forcing due to anthropogenic sulfate.

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